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In the present work, we extended the results of our earlier research in the realm of quantum optics in order to solve the challenging technical problems of importance for the war fighter and homeland security. Specifically, we conducted a broad theoretical and experimental study of new methods of generation of coherence and explored new forms of coherence which were found in order to develop technology in areas, where we have unique expertise, to solve current problems associated with national security and need. For example, we further developed our FAST CARS techniques and standoff detection schemes based on superradiance to allow for the possibility of detecting biochemical pathogens and explosives in the air via real time stand off spectroscopy. We also explored more efficient methods of generation XUV based on quantum coherence.

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Final Report

Contract Number	N00014-08-1-0948
Title of Research	Laser and Stand-off Spectroscopy, Quantum and Statistical Optics
Principal Investigator	Marlan O. Scully
Organization	Texas A&M University, Department of Physics

Goal:

In the present work, we extended the results of our earlier research in the realm of quantum optics in order to solve the challenging technical problems of importance for the war fighter and homeland security. Specifically, we were to conduct a broad theorctical and experimental study of new methods of generation of coherence and explore the new forms of coherence which were found in order to develop technology in areas, where we have unique expertise, to solve current problems associated with national security and need. For example, we were to further FAST CARS techniques, develope standoff detection based on superradiance that would allow for the detection of biochemical pathogens and explosives in the air via real time stand off spectroscopy and develope efficient methods of generation XUV based on quantum coherence.

Accomplishments:

We utilized new understandings of physics and made breakthroughs on the detection of trace gases and biological molecules, extended the limits of CARS spectroscopy, and developed new radiation sources as well as continued investigation into new methods of generation of coherence and exploration the new forms found in order to develop technology in areas, where we have unique expertise, to solve current problems associated with national security and need. The following pages summarize our accomplishments:

II. Activity

B. Research: Current Activities

We continued research into new approaches to a broad range of problems, for example, detection of biothreats such as anthrax and viral agents via FAST CARS technology as developed in our laboratories and standoff detection based on gain-swept superradiance spectroscopy that will allow us to detect biochemical pathogens and explosives in the air and on the ground via real time standing off spectroscopy. In addition, we continued investigations into the fundamentals of laser physics, including methods for generating XUV based on quantum coherence, investigations into of cooperative spontaneous emission and the collective Lamb Shift, Electromagnetically Induced Transparency (EIT), and other various fundamental processes. Specific detail of work done is given below.

1. FAST CARS Work and Stand-off Spectroscopy

- We demonstrated two aspects of the stand-off sensing technique using a femtosecond laser system and an organic dye. First, three cells of a solution of Rhodamine B dye in ethanol were pumped with a single wavelength. Amplification of the light generated by two-photon absorption occurred when the pulses were overlapped temporally, in a direction opposite to that of the pump laser beam. Second, the dispersion of the dye solution permitted two pulses of different wavelength to overlap at a specified location in the solution. The overlapped pulses generated light as the result of two-photon absorption in both the forward and backward directions, relative to that of the pump laser beam.

- We demonstrated a real-time method of measuring the vibrational Raman spectrum of whole blood. Using a novel coherent Raman technique, we recorded the vibrational spectrum of the red blood cells from picoliters in blood in milliseconds. This method could allow for real-time *in vivo* blood monitoring.

- Spatial orientation of molecules is a pervasive issue in chemical physics and, by breaking inversion symmetry, has major consequences in nonlinear optics. We proposed and analyzed an approach to molecular orientation. This extracts from an ensemble of aligned diatomic molecules (equally AB and BA, relative to the E vector) a subensemble that is oriented (mostly AB or BA). Subjecting an aligned molecule to a tailored infrared (IR) laser pulse creates a pair of coherent wave packets that correlate vibrational phase with the AB or BA orientation. Subsequent, suitably phased ultraviolet (UV) or visible pulses dissociate one of these vibrational wave packets, thereby "weeding out" either AB or BA but leaving intact the other orientation. Molecular orientation has significant implications for coherent Raman spectroscopy. In the absence of orientation, coherence between vibrational levels is generated by a pair of laser pulses off which a probe pulse is scattered to produce a signal. Orientation allows direct one-photon IR excitation to achieve (in principle) maximal Raman coherence.
- We studied dynamics of a two-level system driven by a strong off-resonant electromagnetic field. We derived an analytical solution for arbitrary pulse shape. We explored possible applications and tested experimentally as appropriate.
- We demonstrated a femtosecond-oscillator-based system for coherent anti-Stokes/Stokes Raman scattering microscopy, wherein impulsive Raman excitation is combined with narrowband, time-delayed, and therefore, background-free probing. We showed that this simple technique could be used for microscopic imaging with chemical contrast.
- We studied interference between a local oscillator and coherent anti-Stokes Raman scattering signal fields by controlling their relative phase and amplitude. This control allowed direct observation of the real and imaginary components of the third-order nonlinear susceptibility of the sample. In addition, we demonstrated that the heterodyne method can be used to amplify the signal.
- We explored the possibilities of using coherent Raman spectroscopy for real-time detection of biohazards. We examined robustness and definitive means for obtaining a molecule-specific signal to be used in species identification by

exciting vibrational coherence on more than one Raman transition simultaneously. In particular, we concentrated on detecting dipicolinic acid (DPA), known to be a maker molecule for bacterial spores. We considered time-and frequency- resolved techniques for coherent Raman spectroscopy and adapted these for this particular application.

2. Fundamental Laser Physics

Novel Methods of Radiation Generation and optical devices mechanics

- We found that intense short pulses of XUV radiation can be produced by cooperative spontaneous emission from visible or IR laser pulses driving atoms or ions. The process depended on the generation and utilization of atomic coherence as is the case in lasing without inversion. However, the radiation process is not stimulated emission, but is rather cooperative spontaneous emission in the sense of Dicke. We considered the generation of Raman coherence from the perspectives of multi-photon excitation and breaking of adiabaticity and found that the advent of an ultrashort (c.g. single cycle) pulse rewards rethinking of conventional (nanosecond) nonlinear optics with new insights, specifically that many atom mathematics of superradiance has a lot in common with coherent anti-Stokes Raman scattering.
- We experimentally demonstrated an ultra-dispersive optical prism made from a coherently driven Rb atomic vapor. The prism possessed spectral angular dispersion that is 6 orders of magnitude higher than that of a prism made of optical glass; such angular dispersion allows one to spatially resolve light beams with different frequencies separated by a few kilohertz. The prism operated near the resonant frequency of atomic vapor and its dispersion was optically controlled by a coherent driving field.
- We studied the selective reflection of the laser beam from rubidium atomic vapor at the D-2 line (wavelength lambda = 780 nm) at different atomic densities. We used a tunable free-running diode laser. We found a measurable signal at a low atomic density N when the mean distance between resonance atoms reached two

wavelengths. In our experiment, the dimensionless parameter N (1/3) lambda varied from 0.5 to 2.8. The reflectivity increased with density monotonically.

Cooperative Spontaneous Emission (CSE)

- We found that the generation of a new quantum interference effect in spontaneous emission from a resonantly driven system of two identical two-level atoms was due to the spatial variation of the lascr phase at the positions of the atoms. This interference significantly affected the spectral features of the emitted radiation and the quantum entanglement in thc system. The interference lead to dynamic coupling of the populations and coherences in a basis, which was determined by the laser phasc and represented a kind of vacuum mediated super-exchange between the symmetric and antisymmetric states.
- We studied the evolution of timed symmetric N-atom state prepared by conditional absorption of a single photon and exhibiting superradiant decay. We found analytical expression for the initial decay rate of the state valid for any size of spherical atomic cloud. We showed that the timed symmetric state is only approximately an eigenstatc of the system for a large atomic cloud cven if virtual processes are neglected.
- We considered the application of Fermi's golden rule to thc problem of N atoms, one of which was excited, in an extended medium with dimensions that were large compared with the wavelength. We found that, in contrast to an often-voiced opinion, the golden rule does not describe the physics of one photon absorbed and subsequently emitted by N atoms. The correct treatment included many-body effects such as Fano interference. For a finite atomic cloud with size R, in the Markovian limit, the system decayrd exponentially at a rate of around N(λ2/R2) faster than the singlc-atom decay rate.

Lamb Shift

- We analyzed the collective Lamb shift and associated radiative decay of a large cloud of radius R containing N atoms uniformly excited by one photon of wavelength λ. We showed that thc time evolution of the symmetric statc prepared by single photon absorption in the limit $R > \lambda$ is similar to that encountered in the Dicke limit of small sample ($R < \lambda$) superradiance. The theory included virtual (counterrotating) terms naturally and thus provided a simple calculation of the collective Lamb shift of a single Dicke state.

- The problem of single photon collective spontaneous emission, a.k.a. superradiance, from N atoms prepared by a single photon pulse of wave vector $\vec{k}_0$ has been the subject of recent interest. It has been shown that a single photon absorbed uniformly by the N atoms will be followed by spontaneous emission in the same direction. Extending this work we have found a new kind of cavity QED in which the atomic cloud acts as a cavity containing the photon.

- We presented analytical solutions for the evolution of collective state of N atoms. We found a (timed) Dicke state prepared by the conditional absorption of a single photon which exhibited superradiant decay. This is in strong contrast to the evolution of a symmetric Dicke state which is trapped for large atomic clouds. We showed that virtual processes yield only a small effect on the evolution of the rapidly decaying timed Dicke state. However, they change the long time dynamics from exponential decay into a power-law behavior which can be observed experimentally.

Electromagnetically Induced Transparency (EIT)

- We studied the noise spectra in a nonlinear magneto-optical rotation experiment in a rubidium vapor. We observed the reduction of noise in the intensity difference of two orthogonally polarized components of the laser beam. The dependence of the noise level on both the frequency and the longitudinal magnetic field was also studied. We found that the optimal condition for the noise reduction was to work around zero longitudinal magnetic field, where the intensity correlation between the two orthogonally polarized components was at a maximum. Our results could be used to reduce or eliminate the atomic excess noise, therefore improving the sensitivity of nonlinear magneto-optical rotation magnetometers and other atom-optical based applications.

- We experimentally studied the propagation of two optical fields in a dense rubidium (Rb) gas in the case when an additional microwave field is coupled to the hyperfine levels of Rb atoms. Thc Rb energy levels form a close-Lambda three-lcvel system coupled to the optical fields and the microwave field. It has been found that the maximum transmission of the probe field depends on the relative phase between the optical and the microwave fields. We observed both constructive and destructive interferences in electromagnetically induced transparency. A simple theoretical model and a numerical simulation have been developed to explain the observed experimental results.

- We studied a time response of electromagnetically induced transparency (EIT) in a rubidium vapor to a rapid variation of optical phase. Wc found a very fast growth of the absorption when the phase of the optical field was abruptly changed, followed by a slow return to the level of steady-state absorption. Thc recovery time decreased with increasing optical power. A simple theoretical analysis showed that under our experimental conditions the low power limit of the recovery time was determined by the ground relaxation time. In our case it was defined by a time-of-flight of rubidium atoms through laser beam. The obtained value of the ground state relaxation time was in a good agreemcnt with results of direct measurements by 'relaxation in the dark' method. Our technique based on phase dynamics in EIT could be used for investigation of the ground state relaxation and the fast control of EIT.

- We studied electromagnetically induced transparency (EIT) in diatomic cesium molecules in a vapor cell by using tunable diode lasers. We observed a sub-natural resonance in absorption a certain molecular band at different cesium vapor pressures. The width of the EIT resonance showed a linear dependence on a cesium vapor pressure.

Other Various Fundamental Processes

- Wc studied the possibility of creating spatial patterns having subwavelength size by using thc so-callcd dark statcs formed by the interaction between atoms and optical fields. These optical fields had a specified spatial distribution. Our experiments in Rb vapor display spatial patterns that were smaller than the length determined by the diffraction limit of the optical system used in the experiment. This approach may have applications to interference lithography and might be used in coherent Raman spectroscopy to create patterns with subwavelength spatial resolution.

- The time dependent Schrödinger equation is frequently 'derived' by postulating the energy $E \rightarrow i\hbar(\partial/\partial t)$ and momentum $\bar{p} \rightarrow (\hbar/i)\bar{\nabla}$ operator relations. We reviewed the quantum field theoretic route to the Schrödinger wave equation which treats time and space as parameters, not operators. Furthermore, we recalled that a classical (nonlinear) wave equation can be derived from the classical action via Hamiltonian-Jacobi theory. By requiring the wave equation to be linear we arrived at the Schrödinger equation, without postulating operator relations. The underlying philosophy was operational: namely 'a particle is what a particle detector detects.' This led us to a useful physical picture combining the wave (field) and particle paradigms which pointed the way to the time-dependent Schrödinger equation.

- We obtained an analytic solution beyond adiabatic approximation by transferring the 1D Schrodinger equation into the Ricatti equation. Then we showed that our solution was more accurate than JWKB approximation. The generalizations of the approach to 3D are being investigated and possible applications of obtained solutions are being discussed.

- We studied the dipole-dipole spectral broadening of a resonance line at high atomic densities when the self-broadening dominates. The selective reflection spectrum of a weak probe beam from the interface of the cell window and rubidium vapour was recorded in the presence of a far-detuned pump beam. The excitation due to the pump reduced self-broadening. We found that self-broadening reduction dependence on the pump power was atomic density independent. These results provided experimental evidence for the disordered exciton based theory of self-broadening, and could be useful for the description of the interaction of a strong optical field with a dense resonance medium.

- We studied the intensity correlations between two orthogonally linearly polarized components of a laser field propagating through a resonant atomic medium. These experiments were performed in a rubidium atomic vapor. We observed that the correlations between the orthogonally polarized components of the laser beam were maximal in the absence of a magnetic field. The magnitude of the correlations depended on the applied magnetic field. The magnitude first decreased and then increased with increasing magnetic field. Minimal correlations and maximal rotation angles were observed at the same magnetic fields. The width of the correlation function was found to be directly proportional to the excited state lifetime and inversely proportional to the Rabi frequency of laser field.

- Motivated by the recent experimcnt [Sautenkov, V.A.; Rostovtsev, Yu.V.; Scully, M.O. Phys. Rev. A 2005, 72, 065801], we studied the field intensity fluctuations due to interaction between a laser with a finite bandwidth and a dense atomic medium. The intensity-intensity cross-correlation of two orthogonal, circular polarized beams was controlled by the applicd external magnctic field. A smooth transition from perfect correlations to anti-correlations (at zero delay time) of the outgoing beams was observed.

- We revisited the concept of time as it appears in quantum, classical, and statistical physics. By combining special relativity and time-independent quantum field theory, we obtained the time-dependent Schrodinger equation, with time, t, and position, r, being regarded as parameters, not operators. We then examined time as an argument based on the photon wave function in order to show that the time in quantum mechanics is the same as the time in Maxwell's equations. Finally, time was examined from the perspective of statistical time, i.e., time as derived from thermodynamics.

- The dimensional scaling (D-scaling) method first originatcd from quantum chromodynamics by using the spatial dimension D as an order parameter. It later found many useful applications in chemical physics and other fields. It enables, e.g., the calculation of thc energies of the Schrodinger equation with Coulomb potentials without having to solve the partial differential equation (PDE). This is done by imbedding the PDE in a D-dimensional space and by letting D tend to infinity. One can avoid the partial derivatives and then solve instead a reduced-order finite dimensional minimization problem. Nevertheless, mathematical proofs for the D-scaling method remain to be rigorously established. We examined the D-scaling procedures from the variational point of view. Specifically, we showed how the ground state energy of the hydrogen atom model could be calculated by justifying the singular perturbation procedures. In the process, we saw in a more clear and mathematical way how (Herschbach J Chem Phys 85:838, 1986 Sect. II.A) the D-dimensional electron wave function "condenses into a particle".

- We considered collective spontaneous emission from an ensemble of N identical two-level atoms prepared by absorption of a single photon-a.k.a. single photon Dicke superradiance. We found dynamical properties of superradiance for small and large atomic cloud. Moreover, we addressed the effects of virtual processes on collective decay rate and Lamb shift. It turned out that virtual processes lead to relatively small yet interesting effects on the time evolution of a rapidly decaying state. However, such processes substantially modify the dynamics of trapped states by bringing in new channels of decay.

- We presented an experimental and theoretical study of the carrier-envelope phase effects on population transfer between two bound atomic states interacting with intense ultrashort pulses. Radio frequency pulses were used to transfer population among the ground state hyperfine levels in rubidium atoms. These pulses were only a few cycles in duration and have Rabi frequencies of the order of the carrier frequency. The phase difference between the carrier and the envelope of the pulses had a significant effect on the excitation of atomic coherence and population transfer. We developed a theoretical description of this phenomenon using density matrix equations and are exploring the implications and possible applications of these results.

- We studied the dynamics of a two-level system driven by a strong off-resonant electromagnetic field. We derived an analytical solution for arbitrary pulse shape. We discussed possible applications and made an experimental demonstration of the results obtained.
- The fundamental limit to photovoltaic efficiency is widely thought to be radiative recombination which balances radiative absorption. We showed that it is possible to break detailed balance via quantum coherence, as in the case of lasing without inversion and the photo-Carnot quantum heat engine. This yielded, in principle, a quantum limit to photovoltaic operation which can exceed the classical one, and still remain in complete accord with the laws of thermodynamics.

C. Publications

1. FAST CARS Work and Stand-off Spectroscopy

1. G.O. Ariunbold, M.M. Kash, Hebin Li, D. Pestov, V. Sautenkov, Y. Rostovtsev, G.R. Welch, M.O. Scully, "A Model Experiment for Stand-off Sensing", J. Mod. Optics. 55, 3273 (2008).

2. D.A. Goltsov and M.O. Scully, "Real-time monitoring of blood using coherent anti-Stokes Raman spectroscopy," Journal of Biomedical Optics **13**, 054004 (2008).

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2. Fundamental Laser Physics

Novel Methods of Radiation Generation and Optical Devices Mechanics

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Cooperative Spontaneous Emission (CSE)

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Lamb Shift

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Electromagnetically Induced Transparency (EIT)

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Other Various Fundamental Processes

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